

creasing solutions. It is therefore necessary to take a linear combination of the solutions, which corresponds to successive integration along the contours C_2 and C_3 . Deforming these contours and taking into account the analyticity of the integrand, we find that the integration can be carried out along the axis $\eta = 0$, and the main contribution to the integral is made by the vicinities of the points t_6 and t_8 . Accurate to constant factors we obtain as $x \rightarrow -\infty$

$$y \sim t_6^{-\eta} e^{-iW} + t_8^{-\eta} e^{iW} \sim \cos(-\pi/4 + W). \quad (\text{A. 8})$$

For a solution that decreases as $x \rightarrow -\infty$, we must construct contour that passes through the point t_5 . As $x \rightarrow +\infty$, the main contribution to the integration along such a contour is made by the vicinities of the points t_1 and t_4 , which lead to increasing (as $x \rightarrow +\infty$) solutions.

¹⁾ Generally speaking, it will be possible to conclude from (9) that the WKB solution takes the form

$$\exp \left[\pm i \int (k_p^2 + 1/4r^2)^{1/2} dr \right].$$

As shown by Langer^[14] (see, e.g.,^[6]), a consistent allowance for the singularity in the vicinity of $r=0$ leads to the expressions.

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Experimental investigation of spontaneous emission by neon in the presence of a strong monochromatic field

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Quantitative measurements of the spontaneous emission spectra of atoms in the presence of a laser field are performed. Spontaneous emission is recorded for the $3s_2-2p_4$ (0.6328 μ) and $3s_2-2p_{10}$ (0.5434 μ) transitions in neon. The laser field is at resonance with the $3s_2-2p_4$ transition. The line broadening and shifts due to atom-atom and atom-electron collisions are measured. The relative amplitude of the broad spectral component of the nonlinear resonance is determined. The experimental results are used to verify the nonlinear-resonance theory.

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1. INTRODUCTION

The first experimental^[1-4] and theoretical^[5-8] studies of the spectroscopy of spontaneous emission of a gas in the presence of a monochromatic laser field were published relatively long ago. These papers were devoted to the modification of the spontaneous-emission spectrum by the laser field as a result of saturation effects, field splitting, and nonlinear interference effects. These changes manifest themselves as relatively sharp spectral structures (resonances) superimposed on inhomogeneously broadened lines. The shapes of the resonances depend, generally speaking, on the observation

direction, while their widths are as a rule much smaller than the Doppler width.

Field-induced changes in the spectra can be registered by measuring the absorption of a probing laser field, or by observing the spontaneous emission from the perturbed levels. The difficulties connected with the low brightness of the spontaneous emission and with the need for using high-resolution spectral apparatus hinder the progress of research on spontaneous emission. The available data^[1,3] are by way of qualitative reports of the phenomena and cannot claim to offer a quantitative check on the theory. At the same time, the

spontaneous-emission method has an important advantage, since the interpretation of the results on spontaneous emission is in many cases simpler than that of the results on absorption of a sounding field.

The purpose of the present study was to obtain by the spontaneous-emission method quantitative nonlinear-resonance data for the lines $\lambda = 0.6328$ ($3s_2 - 2p_4$) and 0.5434 ($3s_2 - 2p_{10}$) μ of neon. The results were used by us to verify the theory and to determine the width and the shift of the resonances due to collisions with electrons and atoms.

2. EXPERIMENTAL CONDITIONS AND DATA-REDUCTION METHOD

The employed experimental setup and the sequence of the measurements have been described in detail in^[9]. The emission from a stabilized single-mode laser ($\lambda = 0.63 \mu$), modulated at a low frequency (18 Hz), after passing through an inclined semitransparent mirror, was incident on a gas-discharge cell with neon. The spontaneous emission of the neon, propagating in the direction opposite to the laser field, was reflected from the aforementioned mirror and entered the recording section, which consisted of a monochromator crossed with a Fabry-Perot interferometer, and a photomultiplier. The alternating part of the signal was detected, amplified, and recorded with an automatic plotter. Thus, when the interferometer was scanned, the spectrogram recorded only the laser-field-induced modification of the spectrum.

In the observation of the spontaneous emission on the transition $3s_2 - 2p_4$, the signal was appreciably distorted by the scattered laser light. To eliminate the scattered-light signal when working with this transition, we used additional modulation of the current in the discharge cell, at a frequency 600 Hz, and the recording system was correspondingly modified. Observation of the 0.54μ line was carried out in a polarization that coincided with the polarization of the laser beam, while observation of the 0.63μ line was carried out with perpendicular polarization. The discharge cell, cooled with running water, had an inside diameter 3.3 mm and a length 40 cm. The laser-beam diameter in the cell was ≈ 3 mm. The free spectral range Δ_i of the Fabry-Perot interferometer was 2000 MHz. The half-width γ_i of the interferometer apparatus profile was 32 MHz for both $\lambda = 0.63$ and 0.54μ , and was monitored in the course of the operation.

An exact solution of the problem of the weak-field absorption spectrum in the presence of the strong field interacting with the same or with a neighboring transition is given in the Appendix. In first-order approximation in the nonlinear effect, the changes of the spectrum in the case of "backward" observation are due only to changes in the velocity distribution of the populations. From formula (A.14) of the Appendix we obtain for the transition $3s_2 - 2p_{10}$, $\lambda = 0.5434 \mu$ the following expression for the spontaneous-emission line contour

$$J_\mu(\Omega_\mu) \propto \exp\left[-\left(\frac{\Omega_\mu - \Delta_{mi}}{k_\mu \bar{v}}\right)^2\right] \times \left\{ 1 + A \left[1 + \left(\Omega_\mu + \frac{k_\mu}{k} \Omega - 2\Delta_2 \right)^2 \frac{1}{4\Gamma_2^2} \right]^{-1} + AC_2 \exp\left[-\left(\frac{\Omega - \Delta_{mn}}{k\bar{v}}\right)^2\right] \right\}; \quad (1)$$

$$A = \frac{N_n - N_m}{N_m} \frac{k_\mu}{k} \frac{G^2}{\Gamma_2(\Gamma_m + \nu_m)}, \quad 2\Gamma_2 = \Gamma_{mi} + \frac{k_\mu}{k} \Gamma_{mn},$$

$$2\Delta_2 = \Delta_{mi} + \frac{k_\mu}{k} \Delta_{mn},$$

$$\Omega_\mu = \omega_\mu - \omega_{mi}, \quad \Omega = \omega - \omega_{mn}, \quad k_\mu = \omega_\mu/c, \quad k = \omega/c,$$

$$\bar{v} = (2k_B T/m)^{1/2}.$$

Here ω , k and ω_μ , k_μ are the frequencies and wave numbers of the weak and strong fields; Γ_{ij} , Δ_{ij} , and ω_{ij} are the widths, shifts, and frequencies of the i - j transitions; Γ_j is the decay probability of the j level (including the spontaneous and collision-dominated decays); ν_j is the departure frequency for the strong-collision model. The subscripts $i, j = m, n, l$ corresponds to the levels $3s_2$, $2p_4$, and $2p_{10}$, respectively; G^2 is proportional to the strong-field intensity and the observed signal is proportional to A .

The half-width $2\Gamma_2$ of the first term in the square brackets is much smaller than $k_\mu \bar{v}$, i. e., it describes the sharp part of the field structure, or the "peak." The second term in the square brackets with half-width $\sim k_\mu \bar{v}$ will henceforth be called the "band" or the "pedestal." The coefficient C_2 determines the ratio of the amplitudes of the band under the peak at $\Omega = \Delta_{mn}$, and from formula (A.14) we have

$$C_2 = \gamma \pi^{-1} \frac{2\Gamma_2}{k_\mu \bar{v}} \frac{\bar{v}_m + A_{m0}}{\Gamma_m + \nu_m - \bar{v}_m - A_{m0}}, \quad (2)$$

where \bar{v}_m is the arrival frequency and A_{m0} is the Einstein coefficient for the transition to the ground state. If the substitutions

$$k_\mu \rightarrow k, \quad l \rightarrow n, \quad \Delta_{mi} \rightarrow \Delta_{mn}, \quad \Gamma_2 \rightarrow \Gamma_1, \quad \Delta_2 \rightarrow \Delta_1, \quad C_2 \rightarrow C_1$$

are made, expressions (1) and (2) describe the spontaneous emission line for the transition $m-n$, $\lambda = 0.6328 \mu$.¹⁾

We proceed to describe the procedure used to reduce the spectrograms. An important item here is the correct separation of the sharp peak from the pedestal. To this end, a procedure was developed,^[10] in which it was assumed that the overlap of the wings of the pedestal from different interference orders produces a homogeneous background ($\Delta_i \lesssim (0.5-1)k\bar{v}$). Under our conditions $\Delta_i = 2000$ MHz and $k\bar{v} = 825$ and 960 MHz for $\lambda = 0.63$ and 0.54μ , while the line with the Doppler width is marked with a tilde. Failure to take into account the "waviness" of the pedestal overestimates the values of $\Gamma_{1,2}$ by 3-12 MHz and lowers $C_{1,2}$ by a factor of two.

The waviness of the pedestal was taken into account in the following manner. The first approximation was taken to be the value of Γ obtained by the procedure of^[10]. Starting from this value of Γ , with allowance for the overlap of the orders and for the apparatus profiles of the interferometers, formulas (A.11) and (A.14) were used to calculate the amplitude h of the sharp part

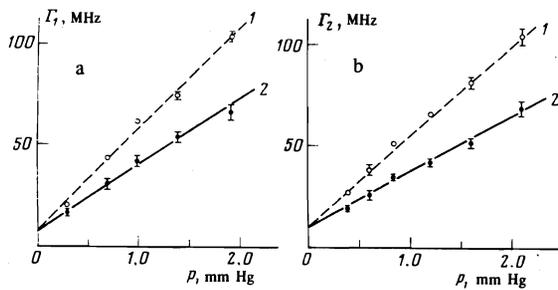


FIG. 1. Pressure broadening of resonance: a) $\lambda = 0.63 \mu$, line 1— $i = 145$ mA, 2— $i = 0$; b) $\lambda = 0.54 \mu$, line 1— $i = 180$ mA, 2— $i = 0$.

at $\Omega_\mu - \Delta_{ml(n)} = \Delta_i/2$. The amplitude h_2 of the pedestal at this point was defined as the difference between the experimental value of the signal and the value of h_1 . The waviness of the pedestal was then calculated from the value of h_1 and under certain assumptions concerning the shape of the pedestal. This was followed by second measurement of the parameters on the spectrogram, with the aid of which the corrected values of Γ and C were obtained by the procedure of [10].

In the calculation of the shape of the pedestal we took into account the absorption of the spontaneous emission as it propagates over the discharge tube. Owing to the self-absorption, the intensity of the radiation that emerges from the tube is proportional to the expression

$$I(\Omega_\mu) \propto \int_0^l J_\mu(\Omega_\mu, x) \exp \left[- \int_0^x \alpha_\mu(\Omega_\mu, x') dx' \right] dx. \quad (3)$$

The dependence of J_μ and of the weak-field absorption coefficient α_μ is due to the absorption by the strong field, the intensity of which is proportional to $\exp[-\alpha(\Omega)x]$, where $\alpha(\Omega)$ is the strong-field absorption coefficient. For $\lambda = 0.63 \mu$, according to these measurements, we have $\alpha(\Omega)l, \alpha_\mu(\Omega_\mu)l \leq 0.4$; in addition, the population N_n of the $2p_4$ level was much higher than that of the $3s_2(N_m)$ level.³⁾ Under these conditions, as shown by analysis, allowance for the self-absorption reduces to multiplying (1) by the factor $1 - \alpha_\mu(\Omega_\mu)l/2$. In view of the large spectral widths of $\alpha_\mu(\Omega_\mu)$, the self-absorption has practically no influence on the contour of the sharp peak in (1) and affects only the shape of the band, a fact also accounted for in the calculation. $\alpha_\mu(\Omega_\mu)$ was then assumed to have a Voigt contour. For the 0.54- μ line, the self-absorption was small and not taken into account.

The line broadening due to the intensity of the laser field (~ 0.2 W/cm²) came into play at pressures p below 1.5 mm Hg, and amounted to 7.0 ± 1.5 MHz at $p = 0.37$

TABLE I.

λ, μ	$\Gamma_{01,2}, \text{ MHz}$	$a_{1,2},$	$b_{1,2},$	$\langle v_e \sigma' \rangle_{1,2}$	$\langle v_e \sigma'' \rangle_{1,2}$
		mm Hg	mm Hg/A		
0.6328	7.7 ± 1.5	32 ± 2	114 ± 14	8.3 ± 1.1	-7.8 ± 0.6
0.5434	10.6 ± 1.5	27 ± 1.5	110 ± 10	7.5 ± 0.8	-8.7 ± 0.8

TABLE II.

Source	$\Gamma_{3s_2}, \text{ MHz}$	$\Gamma_{2p_4}, \text{ MHz}$	$\Gamma_{2p_{10}}, \text{ MHz}$
[12]	—	8.3 ± 0.1	6.4 ± 0.1
[13]	6.4 ± 0.5	6.9 ± 0.6	—
[14]	6.9 ± 0.5	—	—
[15]	—	7.3 ± 1.4	—
[16]	—	6.7 ± 0.4	6.0 ± 0.4
[17]	—	8.15	5.1
[18]	6.4; 6.65; 7.15	9.0; 10.3; 10.3	6.1; 6.95; 7.15
[19]	6.1; 6.4; 6.4	—	—
[20]	8.3 ± 0.3	9.75 ± 0.15	—

mm Hg. The values of $\Gamma_{1,2}$ were extrapolated to zero field intensity. The slight current dependence of the result of the field extrapolation was disregarded. Variation of the discharge current alters, in general, the temperature and the density of the gas. Under the conditions of the present experiments (see^[9]) this factor has practically no influence on the width of the resonance.

Allowance for the aperture broadening resulting from the finite aperture $\Delta\theta$ of the cone of the wave vectors in the registered beam^[6] reduces to subtracting a constant correction $\Delta\theta k\bar{v} = 3$ MHz from $\Gamma_{1,2}$.

Throughout the data reduction it was assumed that $\Omega - \Delta_{mn} = 0$. The experiments were performed in a natural mixture of neon isotopes at pressures p from 0.37 to 2.1 mm Hg and at currents i from 20 to 180 mA.

3. MEASUREMENT RESULTS AND DISCUSSION

The broadening of the resonance by pressure is illustrated in Fig. 1.⁴⁾ The points $\Gamma_{1,2}(i, p)$ (lines 2) were obtained by linear extrapolation to zero currents using four to six values of $\Gamma_{1,2}(i, p)$.

The experimental values of $\Gamma_{1,2}(i, p)$ can be well represented by^[11]

$$\Gamma_\alpha = \Gamma_{0\alpha} + a_\alpha p + b_\alpha p i, \quad \alpha = 1, 2. \quad (4)$$

where Γ_{01} and Γ_{02} are the spontaneous half-widths of the 0.63 and 0.54 μ lines; a_α and b_α do not depend on p or on i . They were determined by us by least squares over the entire aggregate of values of $\Gamma_\alpha(i, p)$ obtained at different pressures and discharge currents, and are listed in Table I.

Tables II and III give the data on the spontaneous width and on the pressure broadening of the $3s_2-2p_4$ transition line of neon and on the widths of the levels $3s_2, 2p_4,$ and $2p_{10}$ obtained by a number of workers and from our results. The most reliable of these are the

TABLE III.

Source	$\Gamma_n, \text{ MHz}$	$\partial\Gamma/\partial p$
[20]	9.05 ± 0.22	—
[21]	8.5 ± 2	—
[22]	15*	25*
[23]	9 ± 1.5	32 ± 2.5
[24]	11±3	24±2
[11]	9.2 ± 1.0	29±2
Present work	7.7 ± 1.5	32±2

*The values were determined from the graph in [21].

values $\Gamma_{2p_4} = 8.3$ MHz,^[13] $\Gamma_{2p_{10}} = 6.4$ MHz^[12] and $\Gamma_{3s_2} = 6.4$ MHz.^[13] Starting from these values we have for the $3s_2-2p_4$ transition $\Gamma_{01} = \Gamma_{0mn} = (\Gamma_{3s_2} + \Gamma_{2p_4})/2 = 7.4$ MHz, which is in good agreement with the measured value of Γ_{01} . The best agreement of the quantity $a_1 = (\partial\Gamma_1/\partial p)_{i=0}$, obtained from the present measurements is with the data of^[11,23].

It is natural to assume that the broadening of the 0.63 and 0.54 μ lines is determined principally by the perturbation of the upper level. Since the 0.63 and 0.54 μ lines have a common upper level ($3s_2$), the collision widths for the transitions $3s_2-2p_4$ and $3s_2-2p_{10}$ should be equal. Under these conditions, the theory yields (see (1))

$$1.02 \leq \Gamma_2/\Gamma_1 \leq 1.08, \quad (5)$$

with the left-hand inequality corresponding to the spontaneous widths and the right-hand inequality to the impact widths. From Table I at $p=1$ mm Hg and $i=100$ mA we have $\Gamma_2/\Gamma_1 = 0.93 \pm 0.15$. The perfectly satisfactory agreement between these numbers can be regarded as an experimental confirmation of the theory of nonlinear resonances.

It is obvious from (4) and (1) that the half-widths Γ_{mn} and Γ_{m1} can be represented in the form

$$\begin{aligned} \Gamma_{mn} &= \Gamma_{0mn} + a_{mn}p + b_{mn}pi, & \Gamma_{m1} &= \Gamma_{0m1} + a_{m1}p + b_{m1}pi, \\ \Gamma_{0mn} &= \Gamma_{01}, & a_{mn} &= a_1, & b_{mn} &= b_1; \\ \Gamma_{0m1} + k_p \Gamma_{0mn}/k &= 2\Gamma_2, & a_{m1} + k_p a_{mn}/k &= 2a_2, \\ & & b_{m1} + k_p b_{mn}/k &= 2b_2. \end{aligned} \quad (6)$$

Using relations (6) and the data of Tables I and II to determine Γ_{0m1} , a_{m1} , and b_{m1} for the $3s_2-2p_{10}$ transition, we obtain

$$\begin{aligned} b_{m1} &= 71 \pm 26 \text{ MHz/A-mm Hg}, \\ \Gamma_{0m1} &= 12.2 \pm 3.4 \text{ MHz}, \\ a_{m1} &= 16.8 \pm 3.8 \text{ MHz/mm Hg}. \end{aligned}$$

The disparity between a_{mn} , b_{mn} (Table I) and a_{m1} , b_{m1} , as well as between Γ_{0m1} and $\Gamma_{0mn} = (\Gamma_{3s_2} + \Gamma_{2p_{10}})/2 = 6.4$ MHz^[12,13] can nevertheless hardly be assigned any importance, since such a recalculation is quite sensitive to the systematic errors of $\Gamma_{1,2}$.

Let us examine the behavior of the quantities $C_{1,2}$. The amplitude of the $C_{1,2}$ pedestal under our conditions is 0.1–0.2 of the amplitude of the peak component. As functions of the pressure, $C_{1,2}$ are practically constant, and decrease with increasing current. These singularities can be readily explained on the basis of the theoretical expression (2) for C_1 and C_2 . Indeed, substituting, for example, the value of Γ_1 from (4) in the expression for C_1 , we obtain

$$C_1 = 2\sqrt{\pi} \frac{\bar{\nu}_m + A_{m0}}{k\bar{\nu}} \frac{\Gamma_{01} + (a_1 + b_1i)p}{\Gamma_m + \nu_m - \bar{\nu}_m - A_{m0} + 2b_1pi} \quad (7)$$

It is assumed in (7) that the electronic broadening is due to inelastic processes, and therefore the probability of the decay of the state m contains the term $2b_1pi$. At pressures $p \sim 1$ mm Hg and currents $i \sim 100$ mA we

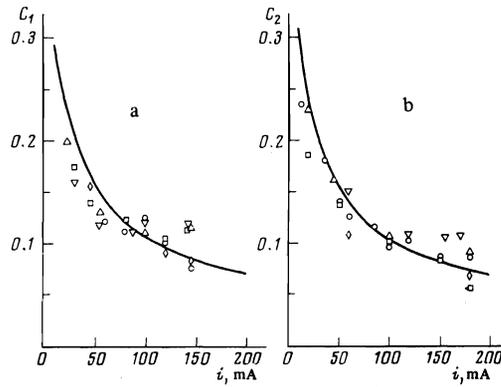


FIG. 2. Ratio of the amplitudes of the $C_{1,2}$ pedestal and peak as functions of the current i and of the pressure p in the discharge: a) $\lambda = 0.63 \mu$, points: \circ — $p = 1.94$ mm Hg, Δ —1.4, ∇ —1.0, \square —0.7, \diamond —0.37; b) $\lambda = 0.54 \mu$; points \circ — $p = 2.1$ mm Hg, Δ —1.6, ∇ —1.2, \square —0.8, \diamond —0.94.

have

$$(a_1 + b_1i)p \gg \Gamma_{01}, \quad 2b_1pi \gg \Gamma_m + \nu_m - \bar{\nu}_m - A_{m0},$$

and consequently C_1 depends little on p . Since, furthermore, $\Gamma_{01} \approx 8$ MHz $>$ $\Gamma_m - A_{m0} \sim 4$ MHz,^[25] it follows that C_1 should decrease with increasing current, as is indeed observed in experiment.

Figure 2 shows a plot of $C_{1,2}(i)$ at $\Gamma_m + \nu_m - \bar{\nu}_m - A_{m0} = 4$ MHz, $\bar{\nu}_m + A_{m0} = 12.6$ MHz, $p = 0.7$ mm Hg and at the values of a_1 and b_1 measured by us. This plot accounts correctly for the experimental behavior of $C_1(i)$, thus providing additional corroboration of the theory.

In a preceding study,^[11] analysis of the spectrum of the stimulated absorption on the transition $3s_2-2p_4$ has revealed an increase of the relative amplitude of the pedestal with increasing current. It was therefore assumed there that an appreciable role in the formation of the pedestal is played by strong collisions (or their analogs) on the $2p_4$ level. Under the conditions of the present study, in contrast to^[11], the relaxation of the lower level does not influence the emission spectrum, and the fact that the relative amplitude behaves qualitatively in a different manner than in^[11] favors the hypothesis advanced there.

The following formula holds for the ratio of the quantities C_1 and C_2 :

$$\frac{C_2}{C_1} = \frac{11}{10} \frac{k \Gamma_2}{k_p \Gamma_1} \quad (8)$$

It was obtained in the strong-collision model from the velocities and the magnetic sublevels. Allowance for the level degeneracy has led to the appearance of a numerical coefficient 11/10 in the right-hand side of (8). At $\Gamma_{mn} = \Gamma_{m1}$ it follows from (8) that $C_2/C_1 = 1.02$. The experimental ratio C_2/C_1 , on the other hand, is 1.0 ± 0.15 , in good agreement with the theory. Using the last value and formula (8), we obtain another independent estimate of the quantity $\Gamma_2/\Gamma_1 = 1.06 \pm 0.16$, which is also in good agreement with (5), i.e., it confirms the theory of nonlinear resonances and the assumption

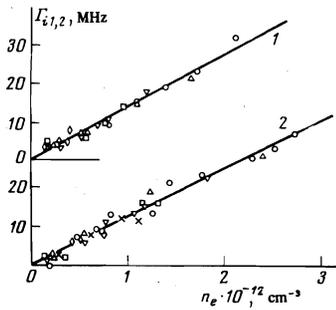


FIG. 3. Current part of the resonance width $\Gamma_{i,2}$ as a function of the electron concentration n_e : 1) $\lambda = 0.63 \mu$, 2) $\lambda = 0.54 \mu$; the symbols for the different pressures in the case of the 0.63 and 0.54 μ lines are the same as in Fig. 2, \times — $p = 0.6$ mm Hg.

that the broadening is due predominantly to the perturbation of the upper $3s_2$ level.

4. LINE BROADENING AND SHIFT BY ELECTRONS

The current part of the broadening and of the shift is due predominantly to collisions with electrons^[9,11]; the half-width and the shift can therefore be represented in the form

$$\begin{aligned} \Gamma_\alpha &= \Gamma_{0\alpha} + \Gamma_{\alpha\alpha} + \Gamma_{i\alpha} = \Gamma_{0\alpha} + a_\alpha p + n_e \langle v_e \sigma_e' \rangle_\alpha, \\ \Delta_\alpha &= \Delta_{0\alpha} + \Delta_{i\alpha} = \Delta_{0\alpha} + n_e \langle v_e \sigma_e'' \rangle_\alpha, \quad \alpha = 1, 2. \end{aligned} \quad (9)$$

Here n_e and v_e are the concentration and velocity of the electrons, σ_e' and σ_e'' are the cross sections for the electronic broadening and shift. We are interested in the quantities $\langle v_e \sigma_e' \rangle_\alpha$ and $\langle v_e \sigma_e'' \rangle_\alpha$. To determine them it is obviously sufficient to know n_e , $\Gamma_{i\alpha}(n_e)$, and $\Delta_{i\alpha}(n_e)$.

The electron concentration n_e is connected with the current i by the relation $i = en_e \bar{v}_d S$, where \bar{v}_d is the average drift velocity of the electrons and S is the cross section area of the tube. The drift velocity \bar{v}_d was determined by us by averaging over all the data given in^[26,27]. The longitudinal electric field intensity \mathcal{E} as a function of i and p , which we need in order to determine \bar{v}_d , was measured by us by a probe procedure. The measured values $\mathcal{E}(i, p) \sim 11-15$ V/cm agree with the data given in^[26]. The dependence of \mathcal{E} on i and p leads to a deviation from the linear $n_e(p, i)$ dependence, thus limiting the accuracy of the representation (4). The obtained values, accurate to $\leq 5\%$, are represented in the form

$$n_e = (11.0 - 2.1p - 3.4i) p i. \quad (10)$$

where n_e is in units of 10^{12} cm^{-3} , p is in mm Hg, and i is in amperes. The scatter in the values of \bar{v}_d obtained by various authors^[26,27] seems to permit the absolute value of n_e to be determined with an error amounting to a factor of 1.5–2.

The values of $\langle v_e \sigma_e' \rangle$ were determined by least-squares reduction using (9) and are listed in Table I. The values of $\Gamma_{0\alpha}$ and a_α , obtained by reduction through the use of n_e , agree well with those obtained by reduction with

the aid of the product ip (4). The error of $\langle v_e \sigma_e' \rangle$ does not include the inaccuracy of n_e . Figure 3 illustrates the $\Gamma_{i\alpha}(n_e)$ dependence.

Let us compare the results known to us on the electronic broadening of the 0.63 μ line of neon. The value obtained in^[29] for the arithmetic mean of the quenching of the $3s_2$ and $2p_4$ levels, which gives the electron broadening due only to inelastic processes, is

$$\begin{aligned} & \frac{1}{2} (\langle v_e \sigma_i \rangle_m + \langle v_e \sigma_i \rangle_n) \\ & = (6.8 \pm 1.2) \cdot 10^{-5} \text{ cm}^3 \text{ sec}^{-1}. \end{aligned}$$

According to^[30] we have $\langle v_e \sigma_i \rangle_m / 2 \approx 5 \cdot 10^{-5} \text{ cm}^3 \text{ sec}^{-1}$. The measurements of^[11] yielded $\langle v_e \sigma_e' \rangle_1 = (6.5 \pm 0.5) \cdot 10^{-5} \text{ cm}^3 \text{ sec}^{-1}$. We note that the data cited and our data are in good agreement, even though different methods were used to obtain them and to determine n_e .

The electronic shift $\Delta_{i\alpha}$ was measured in the following way: With the interferometer continuously scanned, the discharge current is switched at the instant when the next minimum of the signal is recorded, so that the peak that follows the minimum is recorded with a different value of the current. The change of the distance between a given peak and an adjacent neighboring peak is in fact the current shift for the given pair of currents. The shift of the 0.63 μ line, in addition, was measured simultaneously by the method proposed in^[9]. Within the accuracy limit, both methods yielded the same results. The current shift of the 0.63 μ line, measured at $p = 0.7$ mm Hg was $\partial \Delta_{i1} / \partial i = 73 \pm 6$ MHz/A when averaged over nine values. For $\lambda = 0.54 \mu$ and $p = 2.0$ mm Hg we obtained $\partial \Delta_{i2} / \partial i = 232 \pm 16$ MHz/A (32 values). Consequently, after converting p and i into n_e , we obtain directly the values of $\langle v_e \sigma_e'' \rangle_{1,2}$ listed in Table I. From the data of^[31,11] it follows that $\langle v_e \sigma_e'' \rangle_1 = -(6.5 \pm 0.6) \cdot 10^{-5} \text{ cm}^3 \text{ sec}^{-1}$.

In analogy with the widths, assuming equal shifts of the lines of the transitions $m-n$ and $m-l$ ($\Delta_{mn} = \Delta_{ml}$), the theory of nonlinear resonances predicts $\Delta_2 / \Delta_1 = \langle v_e \sigma_e'' \rangle_2 / \langle v_e \sigma_e'' \rangle_1 = 1.08$. From experiment we have

$$\langle v_e \sigma_e'' \rangle_2 / \langle v_e \sigma_e'' \rangle_1 = 1.12 \pm 0.12.$$

For the 0.63 and 0.54 μ lines, the ratio of the electronic broadening and shift, measured under the conditions of our experiments, and likewise independent of the inaccuracy of n_e , is equal to

$$\frac{\langle v_e \sigma_e' \rangle_1}{\langle v_e \sigma_e'' \rangle_1} = -1.06 \pm 0.16, \quad \frac{\langle v_e \sigma_e' \rangle_2}{\langle v_e \sigma_e'' \rangle_2} = -0.86 \pm 0.12.$$

This is much less than predicted by the non-adiabatic theory,^[32] according to which the calculations of^[11] yield $\langle v_e \sigma_e' \rangle_1 / \langle v_e \sigma_e'' \rangle_1 = -2.2$, and is in good agreement with the results of the measurements made by another method in^[11]: $\langle v_e \sigma_e' \rangle_1 / \langle v_e \sigma_e'' \rangle_1 = -1.0 \pm 0.13$.

5. CONCLUSION

Let us examine the agreement between the theory of the nonlinear resonances and experiment. Our present measurements have made it possible to compare with

the theory the ratio of the widths and the shifts of the nonlinear resonances in the $3s_2-2p_4$ and $3s_2-2p_{10}$ transitions, which have a common upper level, and also the ratio of the amplitudes of the "pedestals" for these transitions. These ratios agree with the predictions of the theory (within $\sim 15\%$) so that our results constitute the first quantitative confirmation of the theory of nonlinear resonance in spontaneous emission.

Even the very first theoretical paper^[5-8] called for the shape and width of the nonlinear resonances to be anisotropic, i. e., dependent on the observation direction. The effect of anisotropy was experimentally investigated in^[33] for nonlinear resonance in stimulated emission (the neon transitions $2s_2-2p_1$ and $2s_2-2p_4$) and, in the opinion of the authors of the cited reference, quantitative agreement with the theory was reached.

Other experiments confirm the theory of nonlinear resonances qualitatively. In addition to those listed in the review,^[34] notice should be taken of the experimental studies of the absorption of a weak probing field in the presence of a saturating strong field, in the $3s_2-2p_4$ transition of neon,^[35] which have revealed a strong dependence of the shape and width of the absorption line on the polarization state of the probing and saturating fields.

We emphasize that the most reliable data on the absolute values of $\partial\Gamma/\partial p$, $\langle v_e\sigma_e' \rangle$, and $\langle v_e\sigma_e'' \rangle$, obtained in the neon term system by different methods of nonlinear laser spectroscopy (absorption of a weak field in the presence of a strong one, magnetic scanning, spontaneous emission in the presence of an external field, and others) are all in perfectly good agreement with one another.

It can thus be concluded from the entire aggregate of the presently available experimental data that the theory of nonlinear resonances in the model of relaxation constants and strong collisions has been quantitatively corroborated.

APPENDIX

The problem of the absorption and emission (spontaneous, stimulated) of a probing field in the presence of a strong field (traveling monochromatic wave), for the same transition (two-level system) or an adjacent transition (three-level system), in the model of strong collisions and nondegenerate states, was solved exactly for arbitrary intensity of the strong field.

In a two-level system (transition $m-n$) the matrix element of the interaction is given by

$$V_{mn} = G \exp[-i(\omega t - \mathbf{k}\mathbf{r})] + G_n \exp[-i(\omega_n t - \mathbf{k}_n \mathbf{r})], \quad (A.1)$$

$$G = E d_{mn}/2\hbar, \quad G_n = E_n d_{mn}/2\hbar,$$

where d_{mn} is the dipole-moment matrix elements and E_μ is the amplitude of the weak classical field or the amplitude of the vacuum oscillations at the frequency ω_μ .

The density matrix elements ρ_{ij} in the case (A.1) can

be represented in the form

$$\rho_{jj} = R_{jj} + r_{jj} \exp[-i(\varepsilon t - \mathbf{q}\mathbf{r})] + r_{jj}^* \exp[i(\varepsilon t - \mathbf{q}\mathbf{r})], \quad j=m, n;$$

$$\rho_{mn} = \{R_{mn} + r_{mn} \exp[-i(\varepsilon t - \mathbf{q}\mathbf{r})] + \bar{r}_{mn} \exp[i(\varepsilon t - \mathbf{q}\mathbf{r})]\} \times \exp[-i(\Omega t - \mathbf{k}\mathbf{r})]; \quad (A.2)$$

$$\rho_{nn} = \rho_{nn}^*,$$

where

$$\varepsilon = \Omega_\mu - \Omega, \quad \mathbf{q} = \mathbf{k}_\mu - \mathbf{k}, \quad \Omega_\mu = \omega_\mu - \omega_{mn}.$$

The matrix elements R_{ij} correspond to the solution of the problem with only one strong field G . The increments r_{ij} and \bar{r}_{mn} are due to the weak field G_n .

Under stationary conditions, the values of R_{ij} , r_{ij} , and \bar{r}_{mn} do not depend on the coordinates and time, and satisfy the following matrix equations (in analogy with^[36]):

$$[\hat{\Gamma} - i(\hat{\Omega}' + \hat{G})] \hat{R} - \hat{v} \langle \hat{R} \rangle W(\mathbf{v}) = \hat{Q}, \quad (A.3)$$

$$[\Gamma - i(\varepsilon' + \hat{\Omega}' + \hat{G})] \hat{r} - \hat{v} \langle \hat{r} \rangle W(\mathbf{v}) = i \hat{G}_n \hat{R}. \quad (A.4)$$

The angle brackets denote integration with respect to the velocity \mathbf{v} , and $W(\mathbf{v})$ is the Maxwellian distribution. The matrices in (A.3) and (A.4) have the following structure:

$$\hat{\Gamma} = \begin{pmatrix} \Gamma_m + \nu_m & 0 & 0 & 0 \\ -\gamma_{mn} & \Gamma_n + \nu_n & 0 & 0 \\ 0 & 0 & \Gamma_{mn} + i\Delta_{mn} & 0 \\ 0 & 0 & 0 & \Gamma_{mn} - i\Delta_{mn} \end{pmatrix}, \quad \hat{\Omega}' = \hat{E} \begin{pmatrix} 0 \\ 0 \\ \Omega' \\ -\Omega' \end{pmatrix},$$

$$\hat{\varepsilon}' = \varepsilon' \hat{E}, \quad \hat{v} = \hat{E} \begin{pmatrix} \tilde{\nu}_m + A_{m0} \\ \tilde{\nu}_n \\ \tilde{\nu}_{mn} \\ \tilde{\nu}_{mn}^* \end{pmatrix}, \quad \hat{Q} = \begin{pmatrix} Q_m \\ Q_n \\ 0 \\ 0 \end{pmatrix},$$

$$\hat{R} = \begin{pmatrix} R_{mm} \\ R_{nn} \\ R_{mn} \\ R_{mn}^* \end{pmatrix}, \quad \hat{r} = \begin{pmatrix} r_{mm} \\ r_{nn} \\ r_{mn} \\ \bar{r}_{mn}^* \end{pmatrix}, \quad \hat{G} = \begin{pmatrix} 0 & 0 & G^* & -G \\ 0 & 0 & -G^* & G \\ G & -G & 0 & 0 \\ -G^* & G^* & 0 & 0 \end{pmatrix}, \quad (A.5)$$

$$\hat{G}_\mu = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & G_\mu \\ G_\mu & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad \begin{pmatrix} 0 & 0 & 0 & -G_\mu \\ 0 & 0 & 0 & G_\mu \\ G_\mu & -G_\mu & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},$$

$$\Omega' = \Omega - \mathbf{k}\mathbf{v}, \quad \varepsilon' = \varepsilon - \mathbf{q}\mathbf{v}.$$

Here \hat{E} is the unit matrix; Q_m and Q_n are the rates of excitation of the levels m and n ; $\tilde{\nu}_m$, $\tilde{\nu}_n$, and $\tilde{\nu}_{mn}$ are the "arrival" frequencies; γ_{mn} is the constant of the radiative decay of the level m via the $m-n$ channel.

(A.3) is a closed equation for \hat{R} . Its solution was analyzed in^[36], and here it is only generalized to take into account the spontaneous $m \rightarrow n$ transitions. Equation (A.4) for \hat{r} is of the same form as (A.3). The only difference is that the matrix of the system of equations (A.4) was changed by $i\hat{\varepsilon}'$, and the right-hand side was determined by the solution of Eq. (A.3). The matrix \hat{G}_μ was specified concretely for two problems: the spontaneous emission on the $m-n$ transition (first expression) and the work of a weak external field (stimulated emission minus absorption). In the former case the form of \hat{G}_μ was determined in accordance with the prescription of the classical description of spontaneous emission.^[37]

By virtue of the linearity of the initial equations, in a weak field G_μ , it is easy to generalize the problem in question to include the case of an arbitrary spectral composition for a weak field. Namely, at

$$V_{mn} = G \exp[-i(\omega t - \mathbf{k}_\mu \mathbf{r})] + \sum_\mu G_\mu \exp[-i(\omega_\mu t - \mathbf{k}_\mu \mathbf{r})] \quad (\text{A.6})$$

the solution for ρ_{ij} will be the following:

$$\begin{aligned} \rho_{ij} = & R_{ij} + \sum_\mu \{ r_{ij}^\mu \exp[-i(\varepsilon_\mu t - \mathbf{q}_\mu \mathbf{r})] + \bar{r}_{ij}^{\mu*} \exp[i(\varepsilon_\mu t - \mathbf{q}_\mu \mathbf{r})] \}, \\ & \varepsilon_\mu = \Omega_\mu - \Omega, \quad \mathbf{q}_\mu = \mathbf{k}_\mu - \mathbf{k}, \\ \rho_{mn} = & \left(R_{mn} + \sum_\mu \{ r_{mn}^\mu \exp[-i(\varepsilon_\mu t - \mathbf{q}_\mu \mathbf{r})] \right. \\ & \left. + \bar{r}_{mn}^{\mu*} \exp[i(\varepsilon_\mu t - \mathbf{q}_\mu \mathbf{r})] \right) \exp[-i(\Omega t - \mathbf{k} \mathbf{r})], \end{aligned} \quad (\text{A.7})$$

where each of the sets of r_{mn}^μ , $r_{mn}^{\mu*}$, $r_{mn}^{\mu*}$, $\bar{r}_{mn}^{\mu*}$ satisfies Eq. (A.4).

The solutions of Eqs. (A.3) and (A.4) are

$$\langle \hat{R} \rangle = [\mathcal{E} - \langle \hat{A}^{-1} W(\mathbf{v}) \rangle \hat{\mathcal{V}}]^{-1} \langle \hat{A}^{-1} Q \rangle, \quad (\text{A.8})$$

$$\begin{aligned} \hat{R} = & \hat{A}^{-1} [Q + \hat{\mathcal{V}} W(\mathbf{v}) \langle \hat{R} \rangle], \quad \hat{A} = \hat{\Gamma} - i(\hat{\Omega}' + \hat{G}); \\ \langle \hat{r} \rangle = & [\mathcal{E} - \langle \hat{A}^{-1}(\varepsilon') W(\mathbf{v}) \rangle \hat{\mathcal{V}}]^{-1} \langle \hat{A}^{-1}(\varepsilon') i \hat{G}_\mu \hat{R} \rangle, \\ \hat{r} = & \hat{A}^{-1}(\varepsilon') [i \hat{G}_\mu \hat{R} + \hat{\mathcal{V}} W(\mathbf{v}) \langle \hat{r} \rangle], \end{aligned} \quad (\text{A.9})$$

$$\hat{A}(\varepsilon') = \hat{\Gamma} - i(\hat{\varepsilon}' + \hat{\Omega}' + \hat{G}).$$

The matrix elements of the solution (A.8) for the case $\gamma_{mn} = 0$ have been written out in^[36]. The work of the weak field is determined by the matrix element $\langle r_{mn}^\mu \rangle$. It is easy to obtain for it an explicit expression, but this expression is cumbersome and will not be presented here. We confine ourselves only to the first nonlinear corrections. In addition, we neglect the effect of "phase memory" for the $m-n$ transition ($\bar{\nu} = 0$) and assume, as usual, that $\Gamma_{mn} \ll k\bar{\nu}$. Calculation shows that under these conditions the following formula is valid for the spectral density of the spontaneous emission J_μ in the case of "backward" observation ($\mathbf{k}_\mu = -\mathbf{k}$):

$$\begin{aligned} J_\mu \propto & \left\langle \frac{W(\mathbf{v})}{\Gamma_{mn}^2 + (\Omega' - \Delta_{mn})^2} \left\{ N_m + 2G^2(N_n - N_m) \frac{\Gamma_{mn}}{\Gamma_m + \nu_m} \right. \right. \\ & \left. \left. \times \left[\frac{1}{\Gamma_{mn}^2 + (\Omega' - \Delta_{mn})^2} + \frac{\bar{\nu}_m + A_{m0}}{\Gamma_m + \nu_m - \bar{\nu}_m - A_{m0}} \left\langle \frac{W(\mathbf{v})}{\Gamma_{mn}^2 + (\Omega' - \Delta_{mn})^2} \right\rangle \right] \right\} \right\rangle. \end{aligned} \quad (\text{A.10})$$

The expression in the curly brackets in (A.10) together with the factor $W(\mathbf{v})$ constitutes the velocity distribution of the population of the level m in the presence of the strong field. When the apparatus profile of the interferometer is included, the line contour (A.10) is transformed into

$$\begin{aligned} J_\mu \propto & \frac{1}{\sqrt{\pi} \bar{\nu}} \int_{-\infty}^{\infty} \frac{\exp(-v^2/\bar{\nu}^2) dv}{(\Gamma_{mn} + \gamma_m)^2 + (\Omega_m - \Delta_{mn} + kv)^2} \\ & \times \left\{ N_m + 2G^2(N_n - N_m) \frac{\Gamma_{mn}}{\Gamma_m + \nu_m} \left[\frac{1}{\Gamma_{mn}^2 + (\Omega - \Delta_{mn} - kv)^2} \right. \right. \\ & \left. \left. + \frac{1}{\sqrt{\pi} \bar{\nu}} \frac{\bar{\nu}_m + A_{m0}}{\Gamma_m + \nu_m - \bar{\nu}_m - A_{m0}} \int_{-\infty}^{\infty} \frac{\exp(-v'^2/\bar{\nu}^2) dv'}{\Gamma_{mn}^2 + (\Omega - \Delta_{mn} - kv')^2} \right] \right\}. \end{aligned} \quad (\text{A.11})$$

For a three-level system, which we specify the strong field in the transition $m-n$ ($V_{mn} = G \exp[-i(\omega t - \mathbf{k} \mathbf{r})]$) and the weak field in the transition $m-l$ ($V_{ml} = G_\mu \exp[-i(\omega_\mu t - \mathbf{k}_\mu \mathbf{r})]$). The density-matrix elements

of interest to us have the following dependence on the time and on the coordinate:

$$\rho_{ml} = r_{ml} \exp[-i(\Omega_\mu t - \mathbf{k}_\mu \mathbf{r})], \quad \rho_{nl} = r_{nl} \exp[-i(\varepsilon t - \mathbf{q} \mathbf{r})]. \quad (\text{A.12})$$

In the problem of spontaneous emission in the $n-l$ transition, the quantities r_{ml} and r_{nl} , which are independent of the coordinates and of the time, satisfy a matrix equation analogous to (A.3)

$$\begin{aligned} \hat{A} \hat{r} - \hat{\mathcal{V}} \langle \hat{r} \rangle W(\mathbf{v}) = & Q; \\ \hat{r} = & \begin{pmatrix} r_{ml} \\ r_{nl} \end{pmatrix}, \quad \hat{Q} = iG_\mu \begin{pmatrix} R_{mn} \\ R_{mn}^* \end{pmatrix}, \\ \hat{A} = & \begin{pmatrix} \Gamma_{ml} + i(\Delta_{ml} - \Omega_\mu) & iG \\ iG & \Gamma_{nl} + i(\Delta_{nl} - \varepsilon') \end{pmatrix}, \quad \hat{\mathcal{V}} = \begin{pmatrix} \bar{\nu}_{ml} & 0 \\ 0 & \bar{\nu}_{nl} \end{pmatrix}. \end{aligned} \quad (\text{A.13})$$

Here R_{mn} and R_{mn}^* are taken from the solution of (A.3). The solution of this equation takes the form (A.8), with allowance for the change in the expression for the matrices contained therein. In the approximation where only the first nonlinear corrections are used, we have the following analog of (A.11) (we neglect the phase memory, i. e., $\bar{\nu}_{ml} = \bar{\nu}_{nl} = \bar{\nu}_{mn} = 0$):

$$\begin{aligned} J_\mu \propto & \frac{1}{\sqrt{\pi} \bar{\nu}} \int_{-\infty}^{\infty} \frac{\exp(-v^2/\bar{\nu}^2) dv}{(\Gamma_{ml} + \gamma_m)^2 + (\Omega_\mu - \Delta_{ml} + k_\mu v)^2} \\ & \times \left\{ N_m + 2G^2(N_n - N_m) \frac{\Gamma_{mn}}{\Gamma_m + \nu_m} \left[\frac{1}{\Gamma_{mn}^2 + (\Omega - \Delta_{mn} - kv)^2} \right. \right. \\ & \left. \left. + \frac{1}{\sqrt{\pi} \bar{\nu}} \frac{\bar{\nu}_m + A_{m0}}{\Gamma_m + \nu_m - \bar{\nu}_m - A_{m0}} \int_{-\infty}^{\infty} \frac{\exp(-v'^2/\bar{\nu}^2) dv'}{\Gamma_{mn}^2 + (\Omega - \Delta_{mn} - kv')^2} \right] \right\}. \end{aligned} \quad (\text{A.14})$$

At $\Omega_\mu, \Omega \lesssim k\bar{\nu}$ and $\Gamma_{ml} + \gamma_i \ll k\bar{\nu}$, Eq. (A.14) leads to expression (1).

- 1) Formula (2) corresponds to the model of nondegenerate states. When the degeneracy of the levels $3s_2$, $2p_4$ and $2p_{10}$ is taken into account for the indicated polarizations the right-hand sides in (2) are multiplied by the coefficients 10/9 and 100/99 for the 0.54 and 0.63 μ lines, respectively.
- 2) In place of formula (7) of^[10] (the right-hand of which must additionally be divided by 2π), we used for the determination of $C_{1,2}$ more exact expressions obtained by starting from (A.11) and (A.14).
- 3) An estimate based on the measurement of the ratio of the integrated intensities of the spontaneous emission in the $m-n$ transition with and without a laser field yields for typical experimental conditions $N_n/N_m \approx 6$.
- 4) The values $\Gamma_2(i, p)$, $\Gamma_2(i=0, p)$ at $p=0.6$ mm Hg were obtained with an interferometer having a free spectral range $\Delta_i = 500$ MHz, without introducing a correction for the waviness of the pedestal.

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Penning ionization by nonmetastable atoms

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Penning ionization by highly excited atoms is considered. The asymptotic two-center electron wave function is set up for an arbitrary ion+atom system. This function is needed for the solution of many atomic-collision-theory problems involving nonresonant transitions of the electron from one center to another. The results of the calculations are applied to the case of ionization of noble-gas atoms and hydrogen molecules by highly excited helium atoms. It is shown that the process proceeds mainly via the exchange channel. The conclusions of the theory are in good agreement with the experimental results.

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1. INTRODUCTION FORMULATION OF PROBLEM

In gas-discharge physics, besides the Penning ionization process^[1] (ionization by metastable atoms), great interest attaches also to ionization by more strongly excited atoms. In experiments with ionization chambers,^[2-5] for example, an increase of the ionization current due to one fast α particle was observed in gas mixtures in which ionization by metastable atoms is energywise impossible in one collision (He+Ne, Ar+Xe etc.). To produce a gas laser in a recombining plasma^[6] it is important to know the cross sections for the ionization of the impurity gas atoms by the excited helium atoms. Results of the direct measurements of the cross section for the ionization of Ne, Ar, Kr, Xe atoms by He ($3'P$, 3^3P , 3^3S , $3'S$) atoms at a collision energy

600 °K were recently reported.^[7]

We consider processes of the type



the excitation energy of the atom A^* being larger than the ionization potential J_X of the atom X. When such particles approach each other, an autoionization state is produced, the decay of which leads to reaction (1). The magnitude of the decay width and the potential energy of the interaction of the produced state depend strongly on the excitation number of the atom A^* , so that the cross sections of the reactions (1) can differ significantly in absolute magnitude and in the character of the dependence on the collision energy for the different excited